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We summarize herein our accomplishments made possible by AFOSR Grant No. 89-0108. In particular, we carried out studies of the structure and dynamics of Si, Ge, and SiGe surfaces and interfaces, using isothermal molecular dynamics and isobaric-isothermal Monte Carlo techniques. Concomitantly, we calculated the fundamental interactions of fluorine atoms with silicon using ab initio quantum mechanics, in order to understand the mechanism of etching of silicon by fluorine. Fundamental results from those studies suggest that the surface is highly disordered during etching and that this disorder is produced from the energy released during the extremely exothermic reaction of F atoms with silicon. In order to understand oxidation and nitridation of silicon by, e.g., H<sub>2</sub>O and NH<sub>3</sub>, we must begin by determining the nature of the interaction of H atoms with Si, since they are generated during these reactions and desorption of H atoms can be rate-limiting. We completed studies of H atom adsorption, diffusion, and desorption, learning about the complicated nature of diffusion and desorption on the Si(100) surface. We also developed two new ab initio electronic structure tools: (i) pseudospectral full configuration interaction, a method for treating electron correlation numerically and (ii) a reaction barrier following method for finding transition states for chemical reactions.

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**Final Report**  
**on**  
**AFOSR Grant No. 89-0108**  
**Oxidation, Nitridation, and Fluorine Etching of Si**  
**(1 November 1988 - 30 April 1992)**

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*June 29, 1992*

During the three and half years of the tenure of this grant, we constructed our research program and carried out a number of important studies, as summarized below. In particular, we wrote microcanonical (constant NVE) and canonical (constant NVT) ensemble MD codes for studying processes on the (100), (111), or (110) faces of any fcc, bcc, or diamond lattices. We also wrote an isobaric-isothermal (NPT) ensemble MC program to study equilibrium properties of SiGe alloys and superlattices. Although true deterministic dynamics is only available in the NVE ensemble, most surface science experiments are carried out with conditions of constant temperature, constant volume, and constant number of particles (NVT). We chose to operate in a mixed NVE/NVT ensemble, where the near surface layers were treated with correct dynamics (NVE) and the bottom layers were treated with the canonical ensemble (NVT) in order to mimic the actual isothermal reaction conditions. The NPT ensemble is important to utilize in cases where the lattice constants of the alloy are unknown (i.e., the volume of the crystal is allowed to fluctuate).

As a first step toward studying adsorbate dynamics, we studied the reconstruction dynamics, equilibrium structure, and short-time dynamical behavior of the bare Si(100) and Ge(100) surfaces.<sup>1,2</sup> In particular, we carried out finite temperature MD simulations using empirical two- and three-body interaction potentials for Si and Ge that have been fit to bulk properties.<sup>3</sup> Our simulations reconciled several seemingly conflicting, temperature-dependent surface science experiments in terms of a dynamical model where buckled and

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symmetric surface dimers interconvert on a subpicosecond time scale via a surface phonon (the dimer buckling vibration). In particular, the structure of the surface observed was shown to depend on the time scale of the probe.<sup>1,2</sup> We were able to explain both long time scale (scanning tunneling microscopy) and short time scale (electron diffraction and photoemission) experiments based on this dynamical model. On a technical note, our mixed NVT/NVE ensemble was found to produce much more ordered surface structures than NVE dynamics did.<sup>2</sup> Thus, it is important to use this mixed ensemble during all dynamical simulations that involve evolution or absorption of heat (NVT required) and where time-dependent information is desired (NVE required).

We performed isothermal-isobaric ensemble (NPT) MC calculations of the structures of Si-Ge alloys<sup>4</sup> and superlattices<sup>5</sup> as a function of composition and temperature. We predicted the equilibrium structures of the bulk alloy, the alloy surface, and Si-Ge/Si and Si-Ge/Ge superlattices. (Note that the superlattice work is still in progress and is expected to be completed this summer.) Since diffusion of atoms in a bulk lattice is extremely difficult, we developed an algorithm which speeds up the equilibration process considerably. We carry out conventional MC random walks, but every 500 steps we attempt to permute the positions of two atoms picked at random. Then we allow the nearest neighbors to relax around atoms that may have switched and then continue MC steps for another 500 moves. We find that the main difference between the bulk structure (which we found to be a random alloy above  $\approx 150$  K) and the surface of an Si-Ge alloy is that, given the opportunity, Ge will segregate to the surface. This occurs because the Ge surface energy is lower than that of Si (i.e., Ge-Ge bonds are weaker). This has important implications for epitaxial growth of such alloys, in that if surface segregation is to be avoided, growth should be carried out at a low enough temperature to kinetically limit this surface segregation. Features of the superlattice simulations include identification of a mechanism for relieving strain at the interface between the alloy and the pure element: the alloy expands or compresses in the direction normal to the interface. These alloy

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simulations are the first step toward studying the structures of analogous systems such as  $\text{SiN}_x/\text{Si}$  and  $\text{SiF}_x/\text{Si}$  interfaces.

Our first forays into studying silicon surface chemistry involved modelling chemisorption of H and F on  $\text{Si}(100)\text{-}2\times 1$  using first principles quantum mechanical calculations on embedded silicon clusters ( $\text{Si}_x\text{H}_y$ ,  $x=2\text{-}9$ ,  $y = 2\text{-}12$ ) chosen to represent truncated pieces of the  $\text{Si}(100)$  surface.<sup>6-9</sup> The embedding procedure, which surrounds the subsurface Si atoms with a bulk-like environment is described elsewhere.<sup>8</sup> We have developed several new theoretical strategies for obtaining reliable adsorption energetics: we use large clusters to obtain analytic gradient-optimized geometries at the generalized valence bond (GVB-PP) level<sup>10</sup> and then map those geometries onto smaller clusters (e.g., two to six silicon atoms) when performing correlation-consistent configuration interaction (CCCI) calculations to obtain bond energies and heats of reaction.<sup>11</sup> Based on the largest CI calculations we can do on the very large clusters (essentially a complete active space (CAS)-CI), we estimated the error incurred by using the smaller clusters to represent the surface to be  $\approx 0.1$  eV.<sup>8</sup> In order to properly model chemisorption, it is necessary and sufficient to describe the local wavefunction accurately; we have shown<sup>8</sup> that choosing the optimal geometry for the smaller cluster to be that for the larger cluster leads qualitatively to the same local surface wavefunction for both clusters.

Since the cluster model of chemisorption normally treats the limit of zero coverage, which is clearly unrealistic for most situations, we also developed a means of estimating binding energies for chemisorption at high coverages.<sup>8</sup> Essentially, this involves evaluating the lateral interactions between coadsorbates. In particular, we calculate the interaction energy (normally nonbonded repulsions) between adsorbates on two small clusters as a function of distance between the clusters. The atoms in these clusters are oriented in such a way so that the dominant contributions to nonbonded repulsions are due to the adsorbates. These repulsive forces are evaluated at the GVB-PP level, which should provide an upper bound on the repulsion energy (since dynamical correlations not included

in this wave function would tend to decrease this repulsion). We are careful to evaluate counterpoise corrections as a function of distance for basis set superposition errors that are often present in weakly interacting systems. Since we calculate this repulsive interaction as a function of distance, we are able to use a criterion of minimal repulsions to establish equilibrium adsorbate-adsorbate distances on the surface.<sup>8</sup>

The above techniques were used to predict heats of adsorption for H atoms in the monohydride phase on Si(100).<sup>6</sup> Understanding hydrogen interactions with Si(100) is a critical first step before undertaking studies of nitridation of silicon by NH<sub>3</sub> or oxidation of silicon by H<sub>2</sub>O, since H atoms are generated in both of those reactions and desorption of hydrogen is in fact a rate-limiting process. We predicted that the Si-H bond strength is  $\approx 3.8$  eV and that the endothermicity for desorption of H<sub>2</sub> from this phase is  $\approx 3$  eV, which is significantly higher than any of the desorption activation energies measured by LITD or TPD.<sup>12,13</sup> Only two ways exist for the measured activation energy to be lower than the endothermicity: either the measured activation energy is an "apparent" activation energy, which is the difference between (at least) two activation energies for competing steps, or the pathway for hydrogen desorption does not involve the species for which the endothermicity was calculated. We first suggested that desorption and diffusion might be those competing steps.<sup>6</sup> Thus, we also calculated activation energies for surface diffusion of H on Si(100)-2x1.<sup>9</sup> Several different pathways for surface diffusion were studied: (i) hopping from one dangling bond (db) to another on the same Si dimer; (ii) hopping from one db to a db on an adjacent Si dimer in the same row; and (iii) hopping from one db to a db on an adjacent Si dimer in the adjacent dimer row. Our multiconfiguration self-consistent-field and CI calculations for these diffusion pathways predict that the barrier is lowest for hopping from one db to a neighboring db on an adjacent dimer in the same dimer row. This makes sense upon reflection. For example, hopping from one db to another on the same dimer involves moving across a bridging transition state that has small overlap with either db, since both db's point away from the bridging position. Hopping from one

db to a db on an adjacent Si dimer in the next row over is bad because the Si atoms between which the H atom is hopping are  $\approx 5.3$  Å apart; thus, the orbital overlap is low again in the transition state. The reason the barrier is lowest for hopping between dimers in the same row (in the  $[1\bar{1}0]$  direction) is because the orbitals between which the H atom is hopping are only 3.84 Å apart. Thus, *there is a preferred direction of diffusion along dimer rows!* Quantitatively, the barrier for this most favorable pathway is predicted to be about 2.1 eV.

Recently, we have carried out calculations of various possible H<sub>2</sub> desorption pathways for both the monohydride H-Si-Si-H and dihydride H-Si-H species on Si(100), as a function of coverage.<sup>14</sup> We find that the barrier (calculated at the CASSCF level) for direct desorption from the monohydride state (a 1,2-elimination reaction) is over 100 kcal/mol and hence is ruled out as a mechanism for desorption of H<sub>2</sub> for submonolayer or monolayer coverages of hydrogen on Si(100). This originally led us to propose as a mechanism hydrogen desorption from two adjacent monohydrides in the same dimer row,<sup>6</sup> but this direct mechanism still suffers from the fact that it will lead to a barrier at least as high as the reaction endothermicity, which we calculated to be<sup>6</sup> higher than the observed barrier. Thus, as mentioned above, a direct mechanism is out of the question. We had suggested, as mentioned above, that the barrier might be the difference of two activation energies of competing steps, desorption and diffusion, but further analysis of the kinetics of such a mechanism reveals that this is not feasible. Thus we were forced to conclude that desorption at monolayer and submonolayer coverages of hydrogen on Si(100) could not be due to desorption from either two H-Si-Si or one H-Si-Si-H groups. The answer came when we examined the desorption activation energy for the dihydride species, SiH<sub>2</sub>, and found that it varied from 60 kcal/mol at low coverage to 43 kcal/mol at 2 ML coverage of hydrogen. As it turns out, the latter value is in good agreement with thermal desorption data for the dihydride desorption state at high coverage.<sup>13</sup> The dihydride state at 2 ML coverage of hydrogen desorbs at a temperature well below that of the monohydride state, presumably because the higher surface coverage induces lateral repulsions (we calculate

them to be 17 kcal/mol at 2 ML coverage) that destabilize  $\text{SiH}_2$  on the surface. Interestingly, the low coverage limit for the activation energy for  $\text{H}_2$  desorption from  $\text{SiH}_2$  is in good agreement with the activation energy observed for desorption of  $\text{H}_2$  at low coverages, which was thought to proceed via the monohydride state. We are now convinced that the desorption of hydrogen at all coverages up to 2 ML proceeds via  $\text{SiH}_2$  and that the difference in activation energies between di- and mono-hydride states is merely due to differential lateral repulsions at different coverages. This is consistent with the observed first order kinetics, the measured activation energies, as well as with laser-induced fluorescence studies by Zare and co-workers<sup>15</sup> which concluded simultaneously with our work that desorption of  $\text{H}_2$  from the mono- and di-hydride states goes through a common intermediate, which they suggest to be  $\text{SiH}_2$ . In their work, they suggested a possible isomerization mechanism whereby  $\text{H-Si-Si-H}$  converts to  $\text{H}_2\text{Si} + \text{Si}$ , in order to produce the  $\text{SiH}_2$  intermediate. Our original proposal was that H diffusion to unsaturated  $\text{HSi}$  defect sites might be the mechanism for formation of  $\text{SiH}_2$  at low coverages (since we calculate the diffusion barrier to be much lower than any desorption barrier, it will not be rate-limiting). This could still be happening, but we have decided to calculate the barrier for their mechanism to see if it is also energetically feasible; this work should be completed this month and then we will be submitting a whole story on hydrogen desorption pathways.<sup>14</sup>

Turning to another area, we have used the above quantum mechanical methods to predict adsorbate structures and energetics of the initial stages of F-atom etching (i.e., F adsorption and reaction) of  $\text{Si}(100)\text{-}2\times 1$  at 0 K.<sup>7,8</sup> We find that one monolayer (1 ML) of F atoms adds easily to this surface without breaking any Si-Si bonds, releasing a large amount of energy ( $\cong 6$  eV/Si-F bond formed). Above 1 ML, it is exothermic to form a surface that is 75%  $\text{SiF}_{(a)}$  and 25 %  $\text{SiF}_{2(a)}$ , but then 2.5 eV endothermic, if carried out sequentially, to form a surface that is half  $\text{SiF}_{(a)}$  and half  $\text{SiF}_{2(a)}$ . Above this coverage (1.5 ML), we find it is very difficult to add more fluorine, because of large repulsions between

the F atoms. This is consistent with both molecular beam studies, which found that the rate of fluorine adsorption decreases dramatically above 1.5 ML,<sup>16</sup> and XPS studies that find a stable phase is formed that is close to 3:1 SiF<sub>(a)</sub> to SiF<sub>2(a)</sub>.<sup>17</sup> Our calculations are the first to provide such thermodynamic data and to predict a specific fluorinated surface structure.

We then used our ab initio CCCI data for F interacting with Si and other F atoms to refit the parameters of an analytic potential function for Si-F due to Stillinger and Weber (SW).<sup>18</sup> First, we compared predictions of the SW empirical potential (which was fit to the only Si-F data available, that of the gas phase SiF<sub>4</sub> series) to all of our ab initio reaction paths.<sup>19,20</sup> We showed that the SW potential yields very large errors (up to 1-4 eV for binding energies) when compared to our cluster results. The SW function is simply too repulsive for nonbonded interactions and is not attractive enough for bonding interactions. Indeed, this explains why the two sets of constant temperature MD simulations using this potential found only saturation of the Si dimer db's; no etching or breaking of dimer bonds was observed.<sup>21,22</sup> Clearly, the SW empirical potential is insufficient, since it does not predict that fluorine etches silicon at room temperature, when in fact it does so spontaneously. However, since the overall morphology seemed correct, we retained almost the same SW functional form and searched for new parameters to fit our ab initio data. We developed a simulated annealing algorithm to sample the 12-parameter space of the Si-F two- and three-body terms in an unbiased manner, using  $\approx 50$  pieces of ab initio data.<sup>19,20</sup> We then programmed the forces for our new potential designed to properly describe the fluorosilyl layer. We then performed the first ab initio-derived dynamics for a surface chemical reaction, selected results of which we now describe.<sup>19,20</sup>

We exposed an equilibrated Si(100)-2x1 surface to a fairly high pressure of F atoms (one F atom was added a few Å above the surface every 62.5 fs, which corresponds to  $p \approx 200$  atm) up to a coverage of 2.3 ML of fluorine. Two sets of NVT/NVE trajectories with identical initial conditions ( $T_{\text{surface}} = 300$  K and the F atoms had thermal velocities corresponding also to 300 K) were followed, one as a control. We attempted to etch Si

with our potential in one trajectory and with the original SW potential in the control trajectory. We did this because the reported MD simulations with the SW potential that did not etch Si were done at  $\approx 20$  atm pressure<sup>18</sup> and we wanted to make sure that any differences in the two trajectories were rigorously due to the change in potential and not the change in pressure. Indeed, we find that the SW potential still does not etch Si even at this pressure that is 10x higher and still only saturates the db's on the Si dimers. By contrast, we find that our potential reacts readily with Si at room temperature, producing SiF, SiF<sub>2</sub>, and SiF<sub>3</sub> on the surface. However, contrary to the 0 K quantum calculations described previously, the heat generated by the formation of 6 eV Si-F bonds leads rapidly to a disordered phase. Further, fluorine atoms attack not only the Si atoms in the surface dimers, but also attack subsurface atoms. At low coverages, we find that the F atoms mostly adsorb onto the Si surface dimer atoms along the db direction ( $\approx 23^\circ$  off the surface normal) and hence are merely saturating the db's. However, even at 0.5 ML of F atoms, we find that SiF<sub>2</sub> forms on the surface as a minority species. As more and more F atoms adsorb onto the surface, we find that the near surface layers become disordered, and we further find, based on our adsorption probability studies discussed below, that this disorder is key for etching.

No mechanistic models of silicon etching have been developed, other than macroscopic kinetic models that lack microscopic details.<sup>23</sup> Our simulations have already produced information that was unavailable from experiments, e.g., that fluorine atoms attack not just the dimers at 1 ML coverage but also subsurface Si atoms and that even at this coverage, the huge heat release per Si-F bond formed leads rapidly to a disordered surface and near-surface structure with many broken Si-Si bonds.<sup>19</sup> As mentioned above, these trajectories also revealed that induced disorder may be key to the etch rate. We pursued these ideas by comparing the reaction probabilities for perfectly ordered overlayers of adsorbed fluorine to those for disordered surfaces.<sup>20</sup> We find that while the adsorption probability for atomic fluorine is unity on the clean Si(100) surface at room temperature, it

is zero on a perfectly ordered 1 ML surface of fluorine (i.e., where the surface is covered with F-Si-Si-F groups). If we instead consider a disordered 1 ML surface that is produced via the above etching simulations, we find that the adsorption probability is significant (0.15). Thus, it appears that ordered overlayers (e.g., repeating units of F-Si-Si-F dimers) inhibit etching dramatically. In other words, *the reaction is dependent on the continual production of more and more dangling bonds for F atoms to attack, which is the case for the disordered surface.* Further, we find that ordered surface structures above 1.25 ML are unstable with respect to highly disordered surfaces. Thus, we have concluded that this reaction proceeds in particular by local heating-induced disorder, where the local heating is due to the highly exothermic formation of Si-F bonds and where disorder catalyzes formation of dangling bonds necessary for the etching to proceed onward.

As it is always the case that one wants to treat more and more realistic and hence more complicated systems, methods development is an important component of our research program. To this end, we are developing new techniques for treating electron correlation and for finding transition states on potential energy surfaces. First, we have derived equations for a direct full configuration interaction algorithm that utilizes the pseudospectral approach.<sup>24</sup> To date, no one has used this numerical technique for CI. We have shown that it improves the scaling of full CI from  $CN^4$  to  $CMN^2$ , where  $C$  is the length of the CI eigenvector,  $N$  is the number of molecular orbitals, and  $M$  is the number of grid points used in the pseudospectral part of the calculation. Since  $M$  scales like  $N$ , we achieved a factor of  $N$  reduction in scaling, which will allow the execution of full CI calculations on larger molecules than has been possible in the past. We have verified the accuracy of the technique by comparing the total energies for conventional (spectral) full CI, pseudospectral full CI, and two hybrid spectral/pseudospectral CI approaches. We find that one particular hybrid method gives total energies that differ on the order of 1 microhartree from the spectral results, while retaining the scaling savings. We are now moving to a more practical objective, namely implementing a similar algorithm for truncated

CI (e.g., multi-reference single and double excitation CI). Second, we have recently developed an entirely new approach to finding saddle points on potential energy surfaces. This new method corresponds to reaction barrier following rather than reaction path following. We have tested this on both empirical and ab initio potential surfaces and find it to be accurate, elegant, and simple: it utilizes the energy gradient only occasionally and never the energy second derivatives (except at the very last to check the rank of the saddle point). Further, it requires no initial guess, which might bias the solution, for the saddle point configuration. Further work on this method is in progress, and an initial paper is in preparation describing the technique.<sup>25</sup>

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### Publications Acknowledging AFOSR

1. P. C. Weakliem, G. W. Smith, and E. A. Carter, "Subpicosecond Interconversion of Buckled and Symmetric Dimers on Si(100)," *Surface Sci. Lett.*, **232**, L219 (1990).
2. C. J. Wu and E. A. Carter, "Adsorption of Hydrogen Atoms on the Si(100)-2x1 Surface: Implications for the H<sub>2</sub> Desorption Mechanism," *Chem. Phys. Lett.*, **185**, 172 (1991).
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10. P. C. Weakliem and E. A. Carter, "Surface Chemical Reactions Studied via Ab Initio-Derived Molecular Dynamics Simulations: Fluorine Etching of Si(100)," *J. Chem. Phys.*, submitted (1992).

**Participating Graduate Students  
and Postdoctoral Fellows**

1. Paul C. Weakliem (Ph.D. 1992) 4 August 1988 - 15 April 1992  
Thesis Title: "Theoretical Study of the Structure and Reaction of Semiconductor (100) Surfaces"
2. Christine J. Wu (candidate for the Ph.D.) 1 January 1989 - present.
3. Todd J. Martinez (candidate for the Ph.D.) 15 June 1990 - present.
4. Lawrence E. Carter (candidate for the Ph.D.) 15 June 1991 - present.
5. Aseem Mehta (B. S. 1991; now in graduate school at Caltech) 15 June 1990 - 20 September 1991.
6. Gregory W. Smith (M.S. 1990) 4 August 1988 - 4 June 1990.  
Thesis Title: "Interactions of Nitric Oxide and Carbon Monoxide with Palladium and Platinum Atoms"
7. Dr. Irina V. Ionova (postdoctoral fellow; Ph.D. 1990) 1 October 1991 - present.  
Thesis Title: "Aggregation of Some Classes of Mathematical Models by Means of Linear Aggregates"

**SEMINARS AND PAPERS PRESENTED  
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*A. Invited Lectures*

- |                   |   |
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| May 19, 1989      | "Theoretical Studies of Semiconductor and Metallic Surface Chemistry," Innovation Recognition Symposium, Union Carbide, South Charleston, West Virginia.  |
| October 30, 1989  | "Reconstruction and Etching of Si(100)," AFOSR Molecular Dynamics Contractors Conference, Captiva Island, Florida.  |
| August 2, 1990    | "Classical and Quantal Simulations of Metallic and Semiconductor Surface Chemistry," American Conference on Theoretical Chemistry, San Diego, California.   |
| October 23, 1990  | "First Principles Prediction of the Initial Mechanism for F-Atom Etching of Si(100)," Chemistry at Surfaces Symposium, Irvine, California.  |
| November 18, 1990 | "Lateral Interactions in the Kinetics of Etching Silicon: Surface Steric Effects on Mechanism," NATO-NSF Advanced Research Workshop on Dynamical Stereochemistry, Santa Cruz, California.   |
| March 13, 1991    | "Combining Ab Initio Quantum Chemistry and Dynamics at Solid Surfaces," plenary lecture at the Sanibel Symposia, St. Augustine, Florida.  |
| April 16, 1991    | "Theoretical Studies of Fluorine Etching of and Hydrogen Adsorption on Si(100)," at the Division of Physical Chemistry Symposium on Silicon Hydride Chemistry and Silicon CVD Mechanisms, 201st American Chemical Society National Meeting, Atlanta, Georgia. |
| June 26, 1991     | "Simulated Annealing: from Potential Fits to Ab Initio Molecular Dynamics," at a Workshop on Computer Simulations in Chemical Physics: Recent Advances and New Directions, Irvine, California.  |
| July 22, 1991     | "Combining Ab Initio Quantum Chemistry and Dynamics at Solid Surfaces," at the NSF/SERC US/UK Bilateral Workshop for Young Chemists, Liverpool, England, United Kingdom.  |
| October 21, 1991  | "Reactions of F and H Atoms with Si(100) and Structures of Si-Ge Alloys," at the AFOSR Surface Chemistry Contractors Conference, Irvine, California.  |
| March 5, 1992     | "Theory of Silicon Etching," at the Gordon Research Conference on the Surface Chemistry of Electronic Materials, Oxnard, California.  |
| April 8, 1992     | "The Effect of Lateral Interactions on Surface Dynamics and Thermochemistry," in the Symposium on Intermolecular Forces at the American Chemical Society National Meeting, San Francisco, California.   |

### *B. Seminars*

- April 24, 1989 "Theoretical Studies of Semiconductor and Metallic Surface Chemistry." Chemical Physics Seminar, University of Southern California, Los Angeles, California.
- June 1, 1989 "Molecular Dynamics Simulation of Silicon Surface Reconstruction," Dow Chemical Company, Midland, Michigan.
- September 5, 1989 "Classical and Quantal Simulations of Metallic and Semiconductor Surface Chemistry." Chemical Engineering Colloquium, Cornell University, Ithaca, New York.
- September 8, 1989 "Theoretical Studies of Reconstruction and Etching of Silicon Surfaces." Chemistry Departmental Colloquium, Brown University, Providence, Rhode Island.
- December 8, 1989 "Simulations of Metallic and Semiconductor Surface Chemistry," Olin Research Center, Cheshire, Connecticut.
- December 11, 1989 "Simulations of Metallic and Semiconductor Surface Chemistry," IBM Thomas J. Watson Research Center, Yorktown Heights, New York.
- January 16, 1990 "Simulation of Reconstruction and Etching of Si(100)," Physical Chemistry Seminar, University of California, Santa Cruz, California.
- February 21, 1990 "Simulation of Reconstruction and Etching of Si(100)," Physical Chemistry Seminar, University of California, Santa Barbara, California.
- February 26, 1990 "Simulation of Metallic and Semiconductor Surface Chemistry," Physical Chemistry Seminar, University of California, Riverside, California.
- August 20, 1990 "Structure and Chemistry at Metal and Semiconductor Surfaces from Theory," IBM Almaden Research Center, San Jose, California.
- September 19, 1990 "Structure and Chemistry of Transition Metal and Semiconductor Surfaces," Physical Chemistry Seminar, University of Illinois, Urbana-Champaign, Illinois.
- September 24, 1990 "Metallic and Semiconductor Surface Chemistry from Theory," Chemistry Department Colloquium, Smith College, Northampton, Massachusetts.
- November 8, 1990 "Structure and Chemistry of Palladium and Silicon Surfaces," Physical Chemistry Seminar, Stanford University, Palo Alto, California.
- November 26, 1990 "Structure and Chemistry of Transition Metal and Semiconductor Surfaces from Theory," Chemical Physics Seminar, University of California, Los Angeles, California.

- November 27, 1990 "Structure and Chemistry of Metallic and Semiconductor Surfaces," Physical Chemistry Seminar, University of California, San Diego, California.
- July 22, 1991 "Combining Ab Initio Quantum Chemistry and Dynamics at Solid Surfaces," Physical Chemistry Seminar, Cambridge University, Cambridge, England, United Kingdom.
- August 1, 1991 "Mechanisms of Semiconductor Surface Processes from Theory," Seminar for the Science and Technology Center for Electronic Materials Summer Seminar Series, University of Texas, Austin, Texas.
- September 6, 1991 "Sequential and Simultaneous Ab Initio Molecular Dynamics for Clusters and Surfaces," Chemical Physics Colloquium, University of Colorado, Boulder, Colorado.
- September 12, 1991 "Sequential and Simultaneous Ab Initio Molecular Dynamics for Clusters and Surfaces," Chemistry Department Colloquium, Columbia University, New York City, New York.
- November 1, 1991 "Etching and CVD Chemistry at Silicon Surfaces," Chemical Engineering Seminar, University of California, Los Angeles, California.
- January 21, 1992 "Theoretical Studies of Metallic and Semiconductor Surface Chemistry," Chemistry Department Seminar, California State University at Los Angeles, Los Angeles, California.
- May 11, 1992 "Simulations in Materials Science: Mechanisms for Thin Metal Film Growth and Etching/CVD Chemistry of Silicon," Chemical Research Center Seminar, General Electric Company Research and Development Center, Schenectady, New York.
- May 12, 1992 "Sequential and Simultaneous Ab Initio Molecular Dynamics," Physical Chemistry Seminar, Massachusetts Institute of Technology, Cambridge, Massachusetts.

### *C. Contributed Talks and Presentations*

- October 31, 1988 "Molecular Dynamics Simulations of Processes on Silicon Surfaces," poster at the AFOSR Molecular Dynamics Contractors Conference, Newport Beach, California.
- May 10, 1989 "Molecular Dynamics Simulation of Silicon Surface Reconstruction," poster at the West Coast Theoretical Chemistry Conference, San Jose, California (with Paul Weakliem).
- October 18, 1989 "Fluorine Etching of Silicon Surfaces," talk at the Pacific Conference on Chemistry and Spectroscopy, Pasadena, California (with Christine Wu).

- October 19, 1989 "Molecular Dynamics Simulation of Silicon Surface Reconstruction," talk at the Pacific Conference on Chemistry and Spectroscopy, Pasadena, California (with Paul Weakliem).
- March 1, 1990 "Classical and Quantal Simulation of Reconstruction and F Etching of Si(100)," poster at the 1990 Gordon Research Conference on the Chemistry of Electronic Materials, Ventura, California (with Paul Weakliem and Christine Wu).
- March 14, 1990 "Dynamical Structure of the Reconstructed Si(100) Surface by Molecular Dynamics Simulation," poster at the American Physical Society National Meeting, Anaheim, California (with Paul Weakliem).
- March 24, 1990 "Initial Steps Involved in Fluorine Atom Etching of the Silicon(100)-2x1 Reconstructed Surface," talk at the West Coast Theoretical Chemistry Conference, Salt Lake City, Utah (with Christine Wu).
- March 24, 1990 "Study of the Dynamical Structure of the Reconstructed Si(100) Surface Via Molecular Dynamics Simulation," talk at the West Coast Theoretical Chemistry Conference, Salt Lake City, Utah (with Paul Weakliem).
- August 2, 1990 "Fluorine Atom Etching of the Silicon(100)-2x1 Reconstructed Surface," poster at the American Conference on Theoretical Chemistry, San Diego, California (with Christine Wu).
- August 2, 1990 "Study of the Reconstruction and Structure of Semiconductor (100) Surfaces," poster at the American Conference on Theoretical Chemistry, San Diego, California (with Paul Weakliem).
- February 21, 1991 "Structure and Etching of Group IV Semiconductors by Computer Simulation," talk at the First Annual Surface/Interface Research Meeting of the Southern California AVS, Riverside, California (with Paul Weakliem).
- March 28, 1991 "Hydrogen Adsorption, Diffusion, and Desorption on the Si(100)-2x1 Surface," talk at the West Coast Theoretical Chemistry Conference, Moffett Field, California (with Christine Wu).
- March 28, 1991 "Efficient Algorithms for Correlation-Consistent Configuration Interaction," poster at the West Coast Theoretical Chemistry Conference, Moffett Field, California (with Todd Martinez and Aseem Mehta).
- March 3, 1992 "Theoretical Studies of Hydrogen Adsorption, Diffusion, and Desorption on the Si(100)-2x1 Surface," poster at the Gordon Conference on Surface Chemistry of Electronic Materials, Oxnard, California (with Christine Wu).

- March 5, 1992 "Initial Stages of Fluorine Etching of Si(100) by Molecular Dynamics with a First Principles Many Body Potential," poster at the Gordon Conference on Surface Chemistry of Electronic Materials, Oxnard, California (with Paul Weakliem).
- May 21, 1992 "Monte Carlo Simulations of  $\text{Si}_x\text{Ge}_{1-x}/\text{Si}$  and  $\text{Si}_x\text{Ge}_{1-x}/\text{S}=\text{Ge}$  Superlattice Structures," poster at the 13th Annual West Coast Theoretical Chemistry Conference, Richland, Washington (with Lawrence Carter).
- May 21, 1992 " $\text{H}_2$  Desorption and H Adatom Diffusion on Si(100)-2x1," poster at the 13th Annual West Coast Theoretical Chemistry Conference, Richland, Washington (with Christine Wu).
- May 22, 1992 "Pseudospectral Full Configuration Interaction," talk at the 13th Annual West Coast Theoretical Chemistry Conference, Richland, Washington (with Todd Martinez).
- May 22, 1992 "New Ab Initio Methods for Searching for Transition States," talk at the 13th Annual West Coast Theoretical Chemistry Conference, Richland, Washington (with Irina Ionova).